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Progress was made in the development of theories and computational programs for the following topics: infrared-laser-excited adspecies, including energy and phase relaxation, and desorption; ultraviolet-laser-induced chemical vapor deposition; resonance fluorescence at flat surfaces; photochemistry at structured surfaces, including gratings and thin films; phase-conjugated surfaces; spectroscopy in solid matrices; and nonlinear optical process in polymeric systems.

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ABSTRACT OF OBJECTIVES AND ACCOMPLISHMENTS:

Progress was made in the development of theories and computational programs for the following topics: infrared-laser-excited adspecies, including energy and phase relaxation, and desorption; ultraviolet-laser-induced chemical vapor deposition; resonance fluorescence at flat surfaces; photochemistry at structured surfaces, including gratings and thin films; phase-conjugated surfaces; spectroscopy in solid matrices; and nonlinear optical processes in polymeric systems. This is summarized briefly below.

Infrared-laser-excited adspieces. The following four studies have been carried out: (1) The Born and Markov approximations for phonon relaxation and coherent excitation of adsorbed species are shown to be invalid for CO adsorbed on Ni or Cu (a strongly-bound physisorbed system) but valid for the weakly-bound system Ar on W. (2) A master equation approach which includes electron-hole excitations in the substrate reveals that the probability of finding a laser-driven adsorbed CO molecule on a Cu surface in its first excited vibrational state is 0.03, which is encouraging for experiments on laser-stimulated surface reactions involving CO. (3) The vibrational dephasing rate for OH on SiO_2 is found to be considerably faster than the energy relaxation rate, and the calculated value of 4 ps for the dephasing relaxation time (corresponding to a linewidth of 1.3 cm^{-1}) is in good agreement with experiments. (4) A master equation approach shows that a pulsed laser does not lead to a dramatic increase in the rate of desorption, and in the high-intensity limit, resonant heating and desorption reach the same saturation limit for a pulsed laser and for a continuous-wave laser.

Ultraviolet-laser-induced chemical vapor deposition. Using the Rayleigh hypothesis and determining all components of the electromagnetic field (incident laser, reflected field, image field and surface plasmon field), the dynamics of periodic structured growth of Cd on Si, resulting from photolysis of gaseous $Cd(CH_3)_2$, is calculated in order to explain experimental results.

Resonance fluorescence at flat surfaces. A rigorous quantum theory of atomic resonance fluorescence near a flat metallic surface is derived which allows for polarization-dependent detection involving specific transitions between degenerate substrates.

Photochemistry at structured surfaces. The following two studies have been carried out: (1) The photodissociation of I_2 above a Ag grating surface is calculated semiclassically, where there is an optimal distance of the molecule from the surface at which dissociation is a maximum. The reason for this is that the enhancement due to the surface plasmon field as the molecule is brought closer to the surface is offset by a diminution due to line-broadening effects. (2) Due to cross-coupling into long- and short-range surface plasmons and the different coupling nature between radiations from an incident laser and from the molecular dipole to a corrugated thin metallic film substrate, it is shown that enhanced photoabsorption may be achieved through control of the various film parameters.

Phase-conjugated surfaces. An atom near a phase conjugator behaves quite differently than an atom in empty space or in the vicinity of an ordinary (linear) surface, and it is shown with nonlinear optics that an atom in its ground state can fluoresce if it is sufficiently close to a phase conjugator. This phenomenon opens the door to new and novel types of surface spectroscopy.

Spectroscopy in solid matrices. To describe recent experiments on laser-induced chemical reactions in a $HC\ell$- and $C\ell_2$-doped xenon solid, a semiempirical approach called the diatomics-in-ionic-systems (DIIS) method is developed which accounts for the coupling between ionic and neutral species and charge delocalization among host rare-gas atoms. Calculations are carried out by treating 66 Xe matrix atoms via pairwise interactions, including polarization, with the remaining part of the matrix treated as a continuum, and it is shown that the positive charge in the ionic activated complex is distributed most often between several Xe atoms forming, for example, the $Xe_{12}^{+}C\ell^{-}$, $Xe_{6}^{+}C\ell^{-}$ and $Xe_{2}^{+}C\ell^{-}$ molecules ($Xe_{2}^{+}C\ell^{-}$ is the most stable ionic complex). A calculation of the excitation spectrum of $Xe_{12}C\ell$ is in general agreement with experimental data.

Nonlinear optical processes in polymeric systems. The transient behavior of the optical susceptibility of polydiacetylene-toluene sulfonate induced by an ultrafast pump field has been investigated within a two-level model. The phenomena of optical nutation and optical tristability mediated by phonons and virtual excitons are found, which are shown to be different than in direct-gap bulk semiconductors.

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